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Saharan dust deposition over Mont Blanc (French Alps) during the last 30 years

By M. DE ANGELIS[1] and A. GAUDICHET,[2] [1] *Laboratoire de Glaciologie et Géophysique de l'Environnement, BP 96, 38402 Saint Martin d'Hères, France;* [2] *Laboratoire de Microscopie Analytique Appliquée aux Sciences de la Terre, URA CNRS 1404, Université de Paris XII, Avenue du Général de Gaulle, 94010 Créteil Cedex, France*

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ABSTRACT

Based on continuous and very detailed Al and Ca records along a 70 m long ice core drilled in the Mont Blanc summit area (French Alps, 4270 m asl), continental dust flux at high altitude in the Alps was studied for the past 30 years. From 1955 until 1985, total inputs of 885 and 735 μg cm^{-2} were calculated for Al and Ca respectively. Using mineralogical observations and chemical criteria, the relative importance of the local component was estimated close to 10% for Al and to 30% for Ca continental inputs. It was also possible to point out the predominating influence of soil dust sources located in North Africa, which seems to have significantly increased since the early 1970s with very high inputs occurring after 1980. In 1985, which corresponds to the last complete year of our ice record, the North African influence led to Al and Ca annual deposition rates of 64 and 23 μg cm^{-2} yr^{-1}.

1. Introduction

Arid regions in North Africa contribute to the aeolian dust input on a large part of the Northern Hemisphere, from the Middle East westward to the North American Atlantic coast. Atmospheric investigations based on meteorological data and satellite photos have shown that Saharan outbreaks often travel several thousands of kilometers. Chemical and mineralogical dust composition and atmospheric dust load over the Atlantic Ocean have been well documented (Carder et al., 1986; Coude-Gaussen et al., 1987; Prospero et al., 1981; Prospero and Nees, 1987; Savoie and Prospero, 1980; Talbot et al., 1986). More scattered data are available for the Mediterranean (Bergametti et al., 1989; Ganor and Mamane, 1982; Lefevre et al., 1986; Loÿe-Pilot et al., 1986; Tomadin et al., 1984) and Western Europe (Bücher et al., 1983; Prodi and Fea, 1979; Wheeler, 1985), including a few Alpine stations (Prodi and Fea, 1978). Some of these studies include estimates of aerosol deposition rates, but generally only for particular events and never for periods longer than one year. They have nevertheless provided the basis for a rough estimate of dust inputs from Northwestern Africa, the values of which are surprisingly high (Bergametti et al., 1989; Prodi and Fea, 1979; Wheeler, 1985).

More precise knowledge of Saharan dust inputs requires measurement of the dust deposition rate over various areas and for periods covering several years. This would solve the problems linked to geographical representativeness and possible temporal variability. Permanent snowfields can be considered as geochemical records and provide a good opportunity to study the past tens and even hundreds of years. Due to their position, Alpine areas should provide relevant data on Saharan dust inputs over Western Europe. In the following discussions, the generic term "Saharan" refers mainly to Northwestern Africa (southern regions of Morocco, Algeria, Tunisia and Lybia), which is considered as the dominant dust source for this part of Europe. However, as mean annual temperatures in Europe become largely negative only above 4000 m asl, few firn and ice cores have

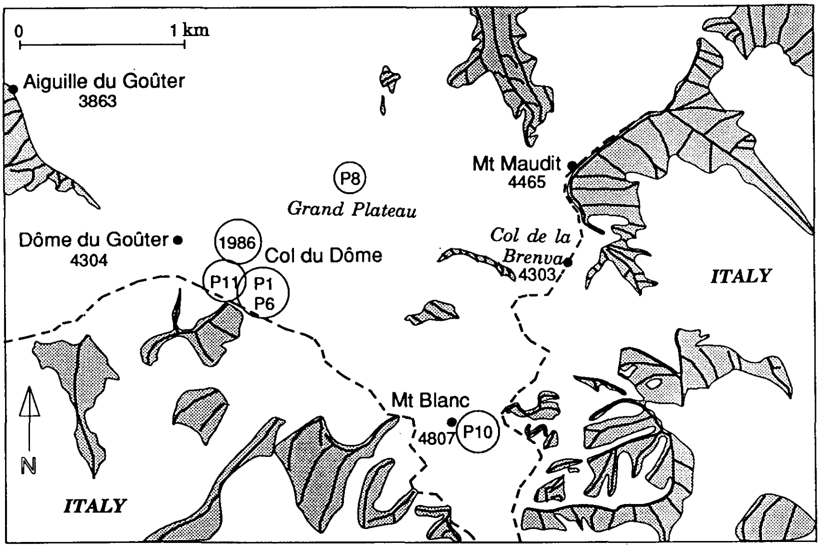

Fig. 1. Map of Mont-Blanc. Sites for which previous snow accumulation data were available are represented by grey circles: P11, Jouzel et al. (1984); P1, P6, P8, P10, Pourchet, personal communication. 1986 corresponds to the drilling site of this work.

been recovered at altitudes high enough to avoid mixing of successive layers by summer melting. A preliminary study published by Briat (1978) concerned anthropogenic and continental impurities included in a 25 m long firn core retrieved at the Col du Dôme (4270 m asl, Mont Blanc) and covering approximately 20 years. More recently, Wagenbach and Geis (1989) calculated a mean Saharan dust deposition rate of 40 μg cm^{-2} yr^{-1} at Colle Gnifetti (4450 m asl, Swiss Alps).

In the beginning of June 1986, a 70 m deep drilling was performed at the Col du Dôme (Fig. 1) by the Laboratoire de Glaciologie et Géophysique de l'Environnement. Based on snow accumulation data previously obtained close to the drilling site by Jouzel et al. (1984), the core was expected to cover about forty years. Our main purpose, when analyzing this core, was to estimate as accurately as possible the relative importance of the Saharan dust contribution to high elevations over the Mont Blanc range. For this, we used major geochemical indicators like Al and Ca, for which the non terrigenous components are known to be negligible at this altitude, and we also performed mineralogical observations on selected samples.

2. Drilling operations

At the Col du Dôme, firn temperature at 19.5 m depth is $-12.4°$C. At this altitude, the thermal balance is almost never influenced by snow melting, and the firn temperature from 10 to 20 m depth is therefore considered to be very close to the mean annual temperature (Jouzel et al., 1984). Although summer air temperature sometimes becomes positive, careful sratigraphic examination of the core attested that percolation remained negligible at this site. By studying a core retrieved less than 100 m far from our drilling site, Jouzel et al. (1984) published for a 10 years time period a mean snow accumulation rate of 109 cm H_2O yr^{-1}, with an annual scattering of 23 cm H_2O yr^{-1}. It must be kept in mind, in the case of solid precipitation, that the measured accumulation rate does not exactly equal the true precipitation depth mainly because of wind activity. During or just after snow fall, part of the precipitation can be blown away in some places by wind erosion, with snow-drifting occurring in other places. However, applying the linear model of Lliboutry (Lliboutry, 1973) to several alpine glaciers (Reynaud, 1977, 1980), it can be shown that the main features of snow redistribution by wind are constant with time at a given location; in other words, although absolute knowledge of snow accumulation rate over any area requires to study the accumulation geographical variability across this area, at a given site and even for time periods no longer than half ten years, the relative changes of annual ice layer thickness observed from year to year are significant. In any case, the drilling site was chosen in an area of weak wind influence and also compressive flow to avoid core break-off by crevasses.

The 69.30 m long core was electromechanically drilled without drilling fluid. Its mean diameter was 90 mm and it seemed to be of very good quality, without any visible fractures.

3. Sampling and analysis

3.1. Sampling

The core was drilled for the purpose of two separate studies, the first concerning Saharan aerosols, the second the anthropogenic influence through ionic balance and heavy metal concentrations. Dating should be based on snow radioactivity and Deuterium variations along the core. All these studies required various sampling procedures leading to rather small aliquots volume for each.

Our sampling rate should have been sufficient to reveal any seasonal or sudden variations of dust concentration but was also depending on firn (or ice) volume available after all the core parcels required by the other studies have been removed and after ultimate cleaning of each core section.

Taking into account previously published accumulation data and preliminary core observations, 10 to 20 cm thick core sections were cut, depending on the depth, so that at least 10 samples per year were obtained.

The close-off, which is the transition from porous firn to air-tight ice, was observed close to 47 m depth. Two procedures were therefore used to remove field (and eventually laboratory) contamination. Below 47.70 m depth, the outer part of each ice section was removed by rinsing with ultrapure water according to the procedure described by De Angelis et al. (1987), except for the part concerning the drilling fluid. From 47.70 m depth up to the surface, the inner part of each core section was melted and sucked through a PTFE Teflon tube. This was done at −2°C under clean room conditions using a thermal probe, 13 mm in diameter, made of chromed steel. It appeared that no percolation occurred during this step, except through the first 2 or 3 mm close to the probe top. Possible contamination, in the ppb range, could have been produced by the apparatus, and was checked for by pouring ultrapure water onto the probe top and sucking it in the same way as for the melted samples. Blank values were 0.6 ± 0.9, 0.3 ± 0.6, 1.1 ± 0.6 and 0.03 ± 0.07 ppb for Al, Ca, Na and K respectively. Confirmation of the cleaning procedure efficiency was obtained for the 10 upper core meters where no samples were required by the other studies, several shallow cores being available. First, the inner part was recored by a system consisting of a PTFE Teflon tube (2 cm in diameter) equipped with 3 molybdenum bits and fastened to the top of a PTFE Teflon box; the complete procedure, described by Batifol (1987) and proved efficient even for trace elements in the ppt range, was unfortunately unsuitable below 10 m depth because of the firn density. After this recoring step, another sampling was made with the thermal probe, close to the first hole, in a similar part of the core as available deeper, i.e. in an externally truncated half-core. Results obtained by these two independent procedures were in very good agreement with respect to all the measured elements and in particular K, which was the most exposed to contamination hasards when drilling or handling the core in the field: two sets of 14 samples with K concentrations varying from 3 to 200 ppb, half being lower than 20 ppb, were studied; while mean overall uncertainty was 14% for the first set of results and 15% for the second one, the slope and the correlation factor of the linear relationship linking the two profiles were respectively 1.09 and 0.995.

3.2. Analysis

A total of 490 successive samples have been obtained. Al, Ca, Na and K were systematically determined by flameless atomic absorption (AA). Aliquots generally ranged between 10 and 50 μl, depending on the considered element, but highly concentrated samples were diluted by a factor of 3 to 10. In each sample, 3 aliquots were systematically analyzed; in cases of analytical scattering greater than 15%, the analysis were repeated 5 to 10 times. Because sample volume available for AA was very small if compared to the size of our PFTE Teflon pressure device and mainly because major sensitivity loss occurred in strongly acidified samples, no digestion step was included in our analytical procedure; however atomisation efficiency was tested by recovery experiments carried out using X-ray fluorescence (XF, 140 samples analyzed for Al) and instrumental neutron activation (INAA, 20 samples analyzed for Al, Ca and Na).

XF was carried out onto 0.4 μm porosity Nuclepore filters, which a few cc's of samples have been passed through, and therefore did not suit for soluble species and probably part of the fine particles. All the samples in the 10 upper meters were analyzed in this way, as were all the samples down to 48 m where concentration peaks have been previously observed by AA. In Fig. 2a it appears that, whatever the scattering around the mean value, AA mean concentrations calculated after 3 to 10 analyses are in very good agreement with XF results (correlation coefficient $r = 0.994$); the slope of the linear regression, AA $= f$(XF), is slightly higher than 1. Moreover, although this linear relationship is partly driven by lowest (<30 ppb) and highest (>3000 ppb) concentrations, its slope and correlation coefficient do not significantly change if only the samples with concentration ranging from 100 to 1500 ppb are taken into account (1.20 instead of 1.11, and 0.972 instead of 0.995). This is a very good basis for concluding that mean values obtained by AA include not only the insoluble component detected by XF, but also an additional component (probably fine and soluble material) corresponding to about 13–17% of the total Al, which is quite plausible (see for instance Prospero and Nees, 1987). Lastly, to account for irregular losses caused by incomplete atomisation, the correlation coefficient should have been lower.

Aliquots of INAA were 1 to 2 cc samples with concentrations ranging from 10 to 4000 ppb and diluted in 20 cc of ultrapure water. INAA allows measurement of the total mass of the element, including both dissolved and suspended matter. Fig. 2b shows that if the point corresponding to the highest concentrations (sample no. 39) is discarded, AA and INAA results are in remarkable agreement ($r = 0.999$) with a linear regression slope of 1.02, proving the validity of AA mean values. The outlying point corresponding to

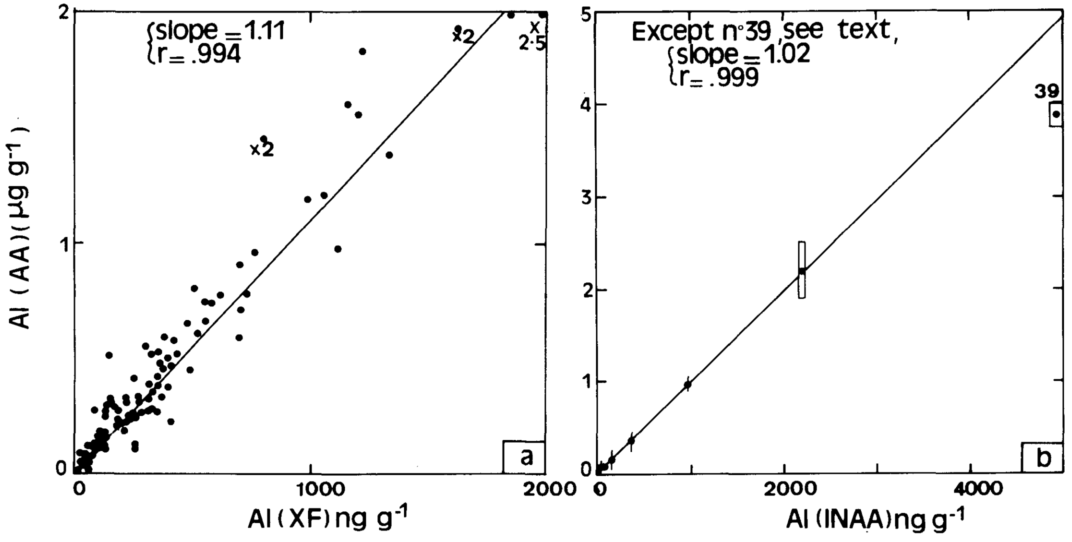

Fig. 2. Comparison between Al concentration mean values obtained by XF (horizontal axis) and AA (vertical axis) is reported in the left part (a). Comparison between Al concentration mean values obtained by INAA (horizontal axis) and AA (vertical axis) is reported in (b), where analytical scattering around each mean value are represented by respectively horizontal and vertical bars.

sample no. 39 could be explained by the presence of few very large particles presumably of local origin. Indeed, concentrations measured in this sample by INAA, AA and XF are 4960, 3860 and 3260 ppb respectively. One to 2 cc aliquots were used for INAA and XF analysis; in the case of AA measurements, 100 µl of samples were diluted in 1000 µl of ultrapure water. If the sample initially contained few particles some ten micrometers in diameter, the aliquot used in the dilution step preceding AA analysis was probably too small to statistically include such particles, which, on another hand should have been heavy enough to have been lost from filter surface before XF (samples being vigorously shaken just before filtering, loss by sedimentation remained always negligible). As indicated in Table 1, see below, traces of small local components were detected in sample no. 39 by mineralogical observations. Fig. 2 refers only to Al results, but recovery between INAA and AA results was also very good for Na (slope 0.95, $r = 0.998$) and, despite the poor sensitivity of INAA for this last element, for Ca concentrations (slope 1.04, $r = 0.998$). Generally speaking, analytical scattering of AA results around the mean values, which are very close to the true ones, could probably be explained by aliquot heterogeneity (and not by atomisation process) as discussed in Subsection 6.1.

Mineralogical studies were performed under a Transmission Electron Microscope (Jeol 100C) fitted with an X-ray Energy Dispersive System (Tracor 420). By filtration of melted snow samples, particles were collected on the surface of Nuclepore membranes (0.2 µm in porosity) previously carbon coated and then transferred directly onto electron microscope gold grids according to a procedure described by Gaudichet et al. (1986). Each grid was scanned at low and higher magnification to estimate, using a squared fluorescent screen, the relative abundance of particles respectively greater than 10 µm, around 5 µm and smaller than 2 µm in projected diameter. Then, mineralogical identification was selectively performed according to these fractional parts. Our main purpose was to identify local or Saharan indicators, taking into account both size and species of the particles. Sixteen samples were studied according to this procedure. Thirteen have been chosen in highly mineralized core levels.

Table 1. *Mineralogical observations and chemical main features of highly concentrated samples*

Ech	Prof	Concentrations				Size distribution			Saharan indicators (fine or mean part.)			Local contribution (large sized part.)	
	(m)	Ca	Δ	Al	Δ	> 10 µm	3–5 µm	< 2 µm	Quartz	Kaol.	Palyg.		
10	1.92	472	1.3	965	8.0	—	+ +	+ +	+ +	+	+ +	no	
35	5.98	238	0.4	742	1.9	—	+	+	+	+	+	no	
39	6.51	636	2.2	3860	3.2	(+)	+	+ +	(+)	(+)	—	low	(angular quartz)
56	8.68	69	2.6	379	16.5		+	+	+ +	+	—	no	
69	10.60	272	40.0	1565	3.1	(+)	—	+ +	—	+ +	—	low	(mica, feldspar)
83	12.94	3098	10.5	5220	10.5	(+)	+	+ +	+ +	+	+	low	(actinote, mica)
85	13.26	1920	38.0	7769	35.0	+	+	+ +	—	(+)	(+)	no	
96	15.23	160	61.0	933	36.0	+ +	—	—	—	—	—	strong	(mica, feldspar, actinote)
125	19.40	160	4.4	7040	15.0	—	+ +	+	+ +	+	—	very low	(actinote)
228	44.81	896	1.9	1600	3.5	+	+ +	+	—	+ +	+ +	low	(actinote)
363	50.92	1455	30.0	2370	33.0	+ + +	—	—	—	—	—	strong	(chlorite)
435	59.40	156	20.0	268	30.0	+ +	—	—	—	—	—	strong	
441	60.17	13400	30.0	348	8.0	+ (very large)	+	+	(+)	—	+	strong	(actinote)

Δ: Relative analytical scattering in %.
—, (+), + and + +: absent, scarse, abundant or very abundant respectively.
Kaol.: kaolinite.
Palyg.: palygorskite fibers.

4. Core dating

Our dating was based on stratigraphic studies and gross-β radioactivity measurements, which were performed on the external part of our samples and allowed to locate 1962 and 1963 summer peaks.

When the core was carefully examined under electric light, large crystal layers, containing regelation ice and considered to have been deposited under warmer conditions (summer and, possibly, late spring or early fall), were observed to alternate with smaller sized crystals layers containing wind-crusts and probably deposited in winter. Such alternation remained rather clear down to 30 m depth and could still be observed deeper in spite of some ambiguity. Then, if Al and Ca profiles, which will be discussed in next paragraph and which consist of peaks alternating with low background periods (see Fig. 3), are compared with stratigraphic data for the upper part of the core, it can be pointed out that summer layers always contain peaks and that extended concentration minima occur in winter. Using winter concentration minima to remove part of the stratigraphic ambiguity and depths of summers 1962 and 1963 deduced from radioactivity measurements (57.5 and 55.8 m respectively, Pourchet personal communication), we were able to propose a chronology summarized in the lower part of Fig. 3. To sum up this chronology, the core bottom should correspond to 1954 or 1955, winter 1969–1970 being located between 47.2 and 49.2 m depth, depending on annual sequences. In Fig. 4, depth is expressed in water equivalent.

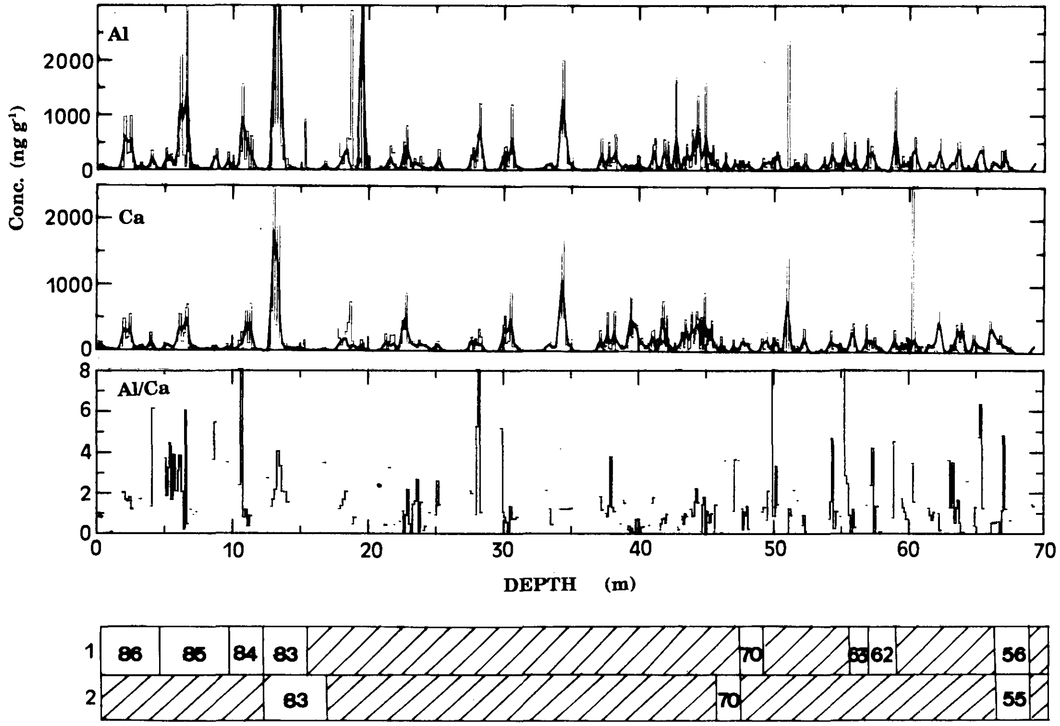

Fig. 3. Col du Dôme (French Alps). Al and Ca concentrations and Al/Ca ratio, calculated for Al and Ca values greater than 60 ppb, as a function of depth; thinner step-shaped lines represent the global component of each continental indicator; thicker lines represent long-range components after smoothing. In the lower part of the figure, the main features of the chronology (1) and an alternative chronology (2) are reported.

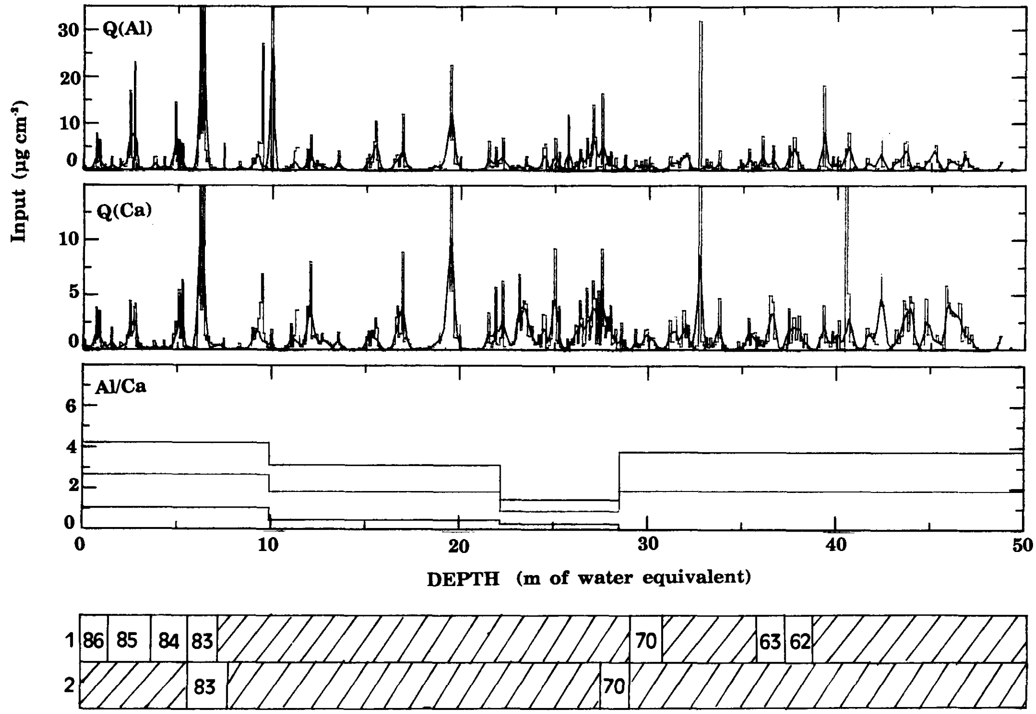

Fig. 4. Col du Dôme (French Alps). Al and Ca inputs as a function of depth expressed in meters of water equivalent; thinner step shaped lines represent long-range components after smoothing. Mean values and scattering of Al/Ca ratio calculated for selected periods are reported as well as the main features of the chronology (see Fig. 3).

5. Global input of Al and Ca

Al is a major component of both the earth's crust and erodible soils. Moreover, its relative abundance does not seem to significantly vary in aerosols over remote continental areas (Lawson and Winchester, 1979) or in various soil categories (Bowen, 1966; Vinogradov, 1959). Thus, it was considered as a suitable continental reference element. Ca is also a major component of most of soils. As high calcium concentrations have often been observed in Saharan aerosol when originating mostly from the North Sahara (Ganor and Mamane, 1982; Hoffman et al., 1977; Lloÿe-Pilot et al., 1986; Schütz and Sebert, 1987), Al to Ca ratio was expected to change with the origin of soil derived dust entrapped in snow and ice layers.

Continuous Al and Ca profiles were established and are presented in part a and b of Fig. 3. A succession of peaks rising up to several hundred and even thousand ppb can be observed. On the other hand, background concentrations vary from sub-ppb values to a few tens of ppb. With a few exceptions, Al and Ca peaks occur in phase, although the Al to Ca ratio significantly varies along the profile. In Fig. 3c, only the ratios corresponding to Al or Ca concentrations higher than 60 ppb have been drawn, more diluted samples being of negligible importance for the following discussion. Beside a few very high values, Al to Ca ratio generally varied between 0.1 and 6. Lowest and less scattered values (0.85 ± 0.59) were measured between 1970 and 1975, while before 1970 and later than 1982, mean values close to 2 with higher scattering were calculated (1.9 ± 1.9 and 2.6 ± 1.6, respectively). Such variations are probably due to change in the relative influence of the different source areas; for example calcite content of the North African source sharply decreases from the coast down to 2000 km southwards (Paquet et al., 1984).

By using density values determined in the field, we calculated individual inputs, $Q_i(\mathrm{Al})$ and $Q_i(\mathrm{Ca})$, corresponding to sample i taken between upper depth U_i and lower depth L_i, of density D_i and concentration C_i:

$$Q_i(\mathrm{Al\ or\ Ca}) = C_i(\mathrm{Al\ or\ Ca})\, D_i(U_i - L_i).$$

Q_i values are reported in Fig. 4 as a function of depth expressed in water equivalent.

By summing Q_i values along the whole profile (except lower and upper layers corresponding to incomplete years), the following total inputs were calculated:

$$Q(\mathrm{Al}) = 885 \pm 170\ \mu\mathrm{g\ cm}^{-2},$$

$$Q(\mathrm{Ca}) = 735 \pm 70\ \mu\mathrm{g\ cm}^{-2}.$$

Considering that the time period covered by this core was between 30 and 31 years, and taking into account the overall uncertainty of Al and Ca inputs, the following values of mean annual continental fluxes were deduced:

$$23 \leqslant \Phi(\mathrm{Al}) \leqslant 35\ \mu\mathrm{g\ cm}^{-2}\ \mathrm{yr}^{-1},$$

$$21 \leqslant \Phi(\mathrm{Ca}) \leqslant 27\ \mu\mathrm{g\ cm}^{-2}\ \mathrm{yr}^{-1}.$$

6. Estimation of the different flux components

Our calculated fluxes correspond to total inputs irrespective of their origin. Above the upper part of Mont Blanc, continental dust probably has two components. The first, we called "long-range" by opposition to "local," originates from mid- or long-range areas (several hundred kilometers) and then travels far from its sources in the middle troposphere; arid areas of North Africa are one of the potential sources of this aerosol component (Bergametti et al., 1989). The second component, which is local, originates from rock outcrops weathered in morainic sands (with scarce vegetation and snow-melt in summer, typically located between 1800 and 3000 m asl) and also from the valleys where soils are significantly remobilized by human activity. In this case, particles can rise up to 4000 m asl only when carried by strong updrafts such as those generated during thunderstorms and local glacier winds.

At first sight, the fact that most of the peaks were observed in warm period layers could be linked to increasing mixing height in summer (at the same time as snow cover is reduced) and consequently to dominant influence of local sources. Thus, any further discussion about continental dust fluxes first required an estimate of the local component and then criteria allowing to distinguish the Saharan input from the remaining long-range component. With this object, we used mineralogical criteria on selected samples in order to check the suitability of chemical criteria available for all the samples. Test results are summed up in Table 1.

6.1. *Local input*

Our first hypothesis was that giant particles ($r > 10\ \mu\mathrm{m}$) should be much more abundant in local than in long-range aerosol (Schütz, 1980). Because of too small available volume, our samples should have been diluted by factor reaching 10 to 20 before Coulter Counter (CC) measurements. Taking into account both blank and statistical problems linked to such high dilutions, we did not use this procedure for particle size distribution. However, several CC studies, performed on polar samples of the glacial age, which contained soil derived particles of long-range origin having log-normal mass distributions centered at 2–4 μm in equivalent diameter (which was rather similar to what Wagenbach and Geis, 1989, observed for Saharan dust), allowed to make assumptions concerning the link between AA analytical scattering and presence or absence of large particles. Indeed, in case of CC measurements, 50 to 500 μl were pumped through the 50 μm probe aperture and, for AA aliquoting, 10 to 50 μl were aspirated through the fine pipette orifice; thus, whatever the analytical procedure, rather similar statistical variations of the number of particles sucked with the melted sample should occur. It appeared from our CC data compared with those of Mounier (1988) that, for a given sample and in each of the 16 CC size ranges, the scattering of the particle number N increased with decreasing N values and could be roughly fitted by a $N^{1/2}$ law when N was lower than a few hundreds. For N values higher than several thousands, the scattering was generally slightly larger, probably because of electronical noise in the first channels and overlapping. Beside these two causes affecting the finest particles, which had a minor effect upon the total particle mass, change in the number of particles aspirated during successive measurements remained the likeliest cause of the observed scattering.

We made a rough estimate of the analytical scattering linked to particle aspiration when 10 μl of a sample containing 500 ppb of Al were sucked, assuming that, in each size range of the particle distribution, the dispersion was equal to $N^{1/2}$. For that to do we used two log-normal distributions measured by CC and for which Al data were available. The first mass distribution was centered at 2 μm, the second at 13 μm. After a linear correction for concentration and volume has been made, we estimated that analytical scattering corresponding to 10 μl successive aliquots should have been 10% in the first case and 35% in the second one. Along the 1986 Mont Blanc core, 64% of the Ca total input (77% for Al) were due to samples with AA analytical scattering lower than 15%, and 32% (22% for Al) to samples with scattering greater than 30%, other samples remaining of minor influence; Wagenbach and Geis (1989) observed Saharan dust distributions with mass mean diameter at 4.5 ± 1.5 μm; thus it was very attractive to assume that samples with analytical scattering greater than 30%, called heterogeneous, corresponded to much larger particles (>10 μm) and then to the local input component. We assessed this first criterion by mineralogical observations on both homogeneous ($\sigma < 15\%$) and heterogeneous samples ($\sigma > 30\%$). Since only a limited number of samples could be studied in this way, the most concentrated samples, by which flux estimations were driven, were observed first. Particles of local origin were characterized using granulometric and morphological criteria (large angular particles greater than 5 μm in diameter) in connection with mineralogical considerations. Mont Blanc is composed of a weathered granitic dome essentially edged with micaschist and amphibolite whereas limestones and shales are dominant in neighbouring valleys. Beside well-known and relatively ubiquitous components like quartz, mica and feldspar, we searched for minerals more typical of local weathered granite and alpine metamorphism like actinote (calcic amphibole) and chlorite, and also for particles typical of carbonated rocks (dolomite and calcite, although calcite could have been partly dissolved). An example of very large chlorite particles is given in Fig. 5a.

It appeared from mineralogical observation of the most concentrated samples that no or few local component was present in homogeneous samples, which allowed us to conclude that the local component was effectively included in heterogeneous samples. Six highly concentrated heterogeneous samples have been studied by electron microscopy. In 5 of these 6 samples, large particles with features corresponding to local origin were observed, they were predominant in 4 of them (sample nos. 96, 363, 435 and 441). These 4 samples represented 90% of the Ca input calculated for the six initial samples (37% for Al) and 80% of the input linked to all the heterogeneous samples (24% for Al). The local input was then calculated by taking into account those 4 samples, proved of local origin, and all heterogeneous samples not subjected to mineralogical observations.

This line of reasoning certainly leads to an overestimate of the local component, since at least one heterogeneous sample has been proved to contain no local features and since, even when significant, the local component does not necessarily account for the whole particle mass. The corresponding results should therefore be considered as upper limits. However minimal inputs were calculated taking into account only the 4 samples proved of local origin by mineralogical controle.

Thus, the following range of local inputs for the last 30 (or 31) years were calculated:

$Q_{\text{local}}(\text{Al}) = 45 - 110\ \mu\text{g cm}^{-2}$
(5–13% of total input),

$Q_{\text{local}}(\text{Ca}) = 215 - 235\ \mu\text{g cm}^{-2}$
(30–32% of total input).

The remaining component should originate from remote areas and has been called the "long-range" input. It accounts for about 85–95% of Al and 70% of Ca global inputs.

6.2. *Saharan input*

The first criterion we thought of to distinguish Saharan from more generally speaking long-range input was chemical. Indeed Saharan aerosol carried far from its source is dominated by quartz and clays (Prodi and Fea, 1979; Tomadin et al., 1984; Wheeler, 1985), which contain no Na.

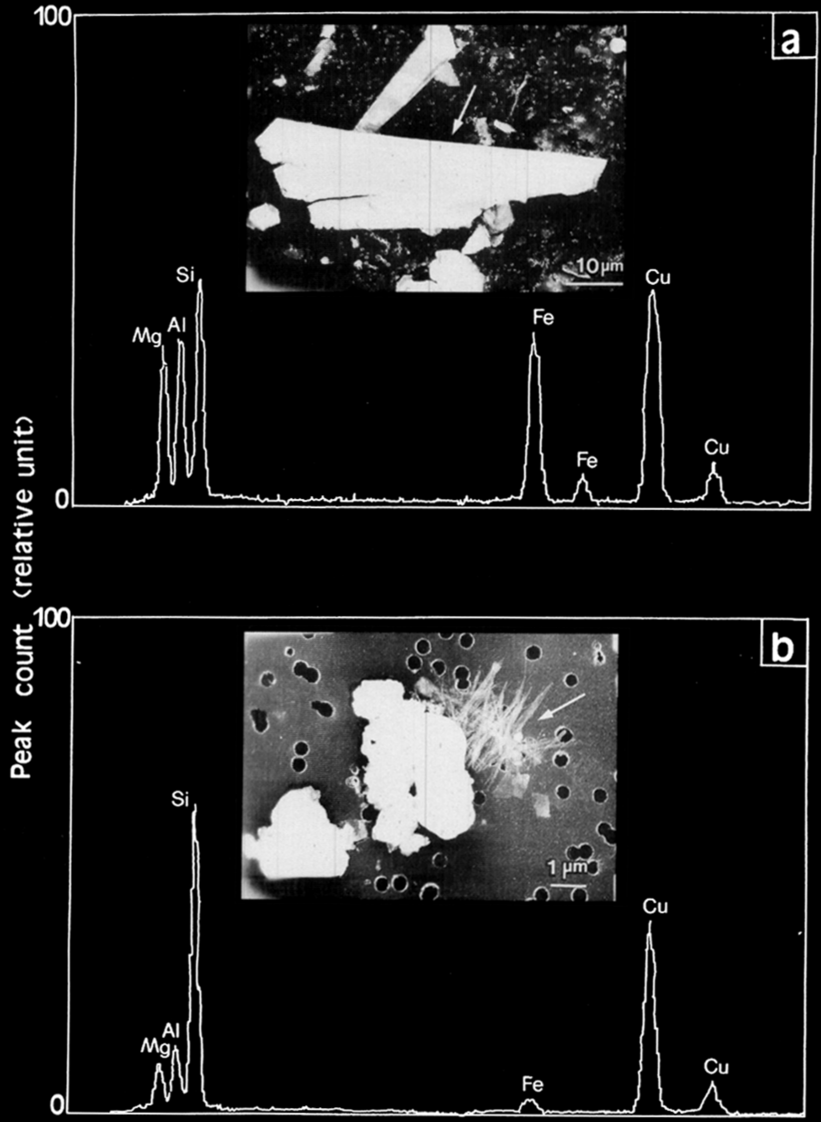

Fig. 5. An example of local input is given in the upper part of the figure (transmission electron micrograph and energy X-ray spectra of large chlorite plates), while an example of Saharan indicator is shown in the lower part (transmission electron micrograph and energy X-ray spectra of thin palygorskite fibers associated with other insoluble soil particles).

Therefore samples in which the local contribution was estimated to be low or negligible and in which K/Na ratio was greater than or equal to 2 were liable to be of Saharan origin. We measured K/Na ratio in all the samples where Al and Ca concentrations were significantly higher than background values, i.e., than 60 ppb, and we checked this criterion using mineralogical observations.

As for local input, we first studied heavily concentrated samples. We looked for mineral associations of at least 2 out of the 3 species generally considered as Saharan origin indicators, i.e. palygorskite individual or gathered fibers (Schütz and Sebert, 1987), rounded quartz and kaolinite (Bücher et al., 1983; Prodi and Fea, 1979; Tomadin et al., 1984; Wheeler, 1985), especially among the fine particles more liable to travel long distances. An example of fine palygorskite fibers is given in Fig. 5b. It seemed futile to search for $CaCO_3$ fine particles, which, even if not completely attacked in the atmosphere by H_2SO_4, would probably have been dissolved when the samples were melted.

Few local particles were observed in the samples studied by electron microscopy, other than the 4 heterogeneous samples proved to be of local origin (see Subsection 6.1). On the contrary, particles were predominantly fine or middle sized, and Saharan indicator associations were observed in all of them. The clay phase was dominated by illite, which, although ubiquitous, is also the major Saharan clay and should have led to high K/Na ratios. In all these samples, where particles were therefore very likely of Saharan origin, K/Na ratio was greater than or equal to 2.

We have finally come to the conclusion that our chemical criterion was valid, although privileging clay rich samples over those in which calcite was the most abundant mineral, and therefore likely leading to an underestimate of the Ca Saharan input.

For the last 30 (or 31) years, Saharan inputs (with corresponding analytical uncertainties) were estimated as:

$$Q_{\text{Sahara}}(\text{Al}) = 454 \pm 35\ \mu\text{g cm}^{-2}$$
(51% of total input),

$$Q_{\text{Sahara}}(\text{Ca}) = 161 \pm 8\ \mu\text{g cm}^{-2}$$
(22% of total input).

41% (for Ca) and 58% (for Al) of those quantities have been controlled by electron microscopy; in particular, Saharan origin was dominating in almost all the peaks observed from the surface down to 20 m depth (from 1986 to ca 1981).

For want of further data, especially mineralogical data, nothing precise can be said about the origin of the residual component, which accounts for 35–45% of Al and 40–50% of Ca global inputs. However, the rather high Ca content of this component, 80% in ionic species and not of marine origin (Ronseaux, personal communication), probably not produced by the weathering of local slightly soluble minerals, could be, at least in part, calcite of Saharan origin not taken into account by our chemical criterion.

7. Discussion

Whatever the origin of the residual long-range component, it was obvious from our results that Saharan input at high elevations in the Mont Blanc area was of importance, and we tried to assess the possible reasons for this.

7.1. Meteorological considerations

We considered the upper part of the core, where annual layer thickness and rather clear stratigraphy allowed us to clearly distinguish successive transitions from winter to summer snow, which approximately correspond to spring and fall snow. Although accurate seasonal divisions were not possible, we made the rough assumption that, at this altitude, winter lasted 6 months (November–April), spring May and June, summer July and August and fall September and October. Using individual inputs, calculated for each sample, and stratigraphic data (see Fig. 4), we were able to make the following estimates which are summarized in Fig. 6a and concern Saharan input:

- In 1983, input began in spring, was very strong in summer and stopped at the beginning of fall. None occurred in winter.
- In 1984, strong input occurred in spring and was markedly lower in summer. One small peak was observed in fall and none occurred in winter.
- In 1985, input was maximum in spring but remained significant in summer; minor events occurred in fall.
- Contrary to the other winters, the 1985–1986 winter was marked by strong input. Little but not negligible activity was also noticeable in spring.

Considering North Africa as probably being the main Saharan dust source for Europe, we consulted daily meteorological reports (Bulletins Météorologiques Européens) over the same period; we looked for meteorological situations allowing air masses to travel from North Africa to Europe and occurring at the same time as or just after dust pickup over source areas. For that, atmospheric pressures and wind patterns were examined at the 500 mb pressure level which is close to the Mont Blanc altitude. However, it must be noted that, under conditions suitable for Saharan dust to reach Alpine areas, 500 mb and 700 mb patterns were in agreement. Dust pickup

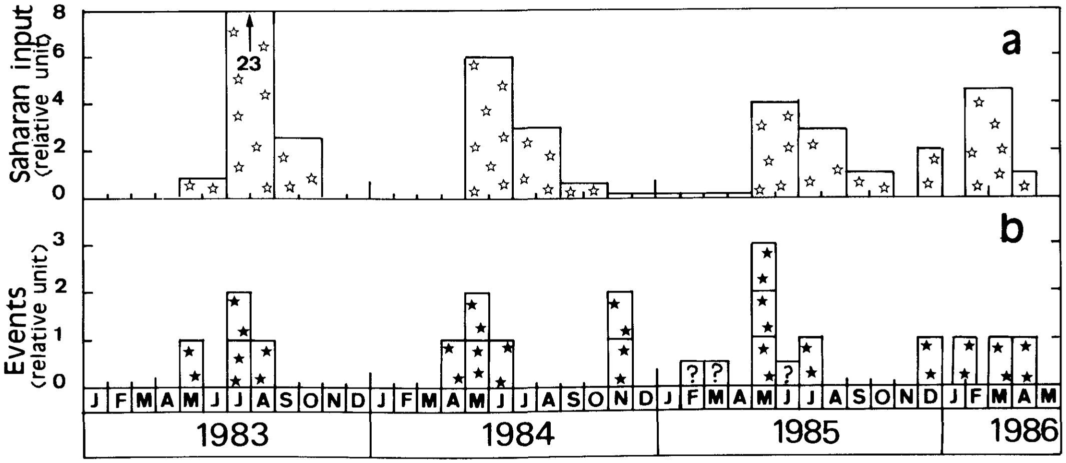

Fig. 6. Seasonal variations of Saharan dust input over the Mont-Blanc upper area (a), and monthly occurrence of meteorological conditions propitious to the transport of North African dust particles over the Alps (b), expressed in relative units.

and transport events lasting at least 2 days were considered to correspond to "suitable meteorological conditions." There are 3 main ways for air masses to travel from North Africa to the Alps: direct and anticlockwise or clockwise, i.e., from the East or West respectively (Prodi and Fea, 1978).

Dust pickups showed no clear spatio-temporal trend except that they were more frequent from April to September. Favorable event occurrences were summed up for each month of the January 1983–May 1986 period, allowing us to plot the lower part of Fig. 6. Taking into account stratigraphic uncertainty, measures performed in snow and meteorological observations were in qualitative agreement, showing major occurrences in summer 1983 and spring 1984 and 1985, and unusual strong input during winter 1985–1986. However nothing significant was observed in snow layers corresponding to the major Saharan outbreak which crossed Western Europe in early November 1984 and led to high dust inputs as far as over England (Wheeler, 1985). Moreover continental inputs in snow are not directly related to our meteorological index. Besides the inaccuracy of this index, it is obvious that, even if atmospheric dust load is increasing, dust flux to snow surface can remain very low, particularly if no precipitation occurs. Indeed, Pourchet et al. (1988) have demonstrated that, in alpine areas, radioactive material injected by the Chernobyl explosion has been removed by wet deposition. If this kind of deposition also prevails for mineral particles and taking into account the spatial variability of precipitation height associated with thunderstorm activity, varying dust quantities should deposit, depending on the site, even in case of homogeneous atmospheric concentrations. For that reason, our feeling is that the impetus of individual events cannot be inferred from our record, which corresponds to mean trends over several weeks or years and allows us rather to calculate and to discuss annual flux mean values.

7.2. *Spatio-temporal comparisons*

Long-range (including Saharan component) and Saharan-only flux components corresponding to various periods have been calculated. Results are presented in Table 2. Mean fluxes calculated over the 1970–1985 period were significantly higher than the ones corresponding to 1955–1969 (by factors of 2.8 and 1.9 concerning the Al and Ca long-range component and by factors of 4.5 and 2.9 concerning the Saharan component of these two elements). This increase became particularly sharp in 1983, since the mean fluxes calculated for the 1983–1985 period, which are almost totally of Saharan origin, are 82 and 34 $\mu g\ cm^{-2}\ yr^{-1}$ for Al and Ca respectively. The 1985 Al flux value, 64 $\mu g\ cm^{-2}\ yr^{-1}$, is slightly lower than the result

Table 2. *Saharan inputs over western Europe*

	Flux $\mu g\ cm^{-2}\ yr^{-1}$				Period	Site	Ref.
	Saharan		Long-range				
**	Ca	Al	Ca	Al			
A	23	64	23	64	1985	Col du Dôme	this work
A	34	82	34	82	1983–1985	—	—
A	7.6	24	21	36	1970–1975	—	—
A	2.6-2.7	5.1-5.5	11-12	13-14	1955(56)–1969	—	—
A	5.2-5.4	16-17	16-17	25-26	1955(56)–1985	—	—
A	2.1-2.2	7.4-7.6	14-15	19-20	1955(56)–1982	—	—
A		3.5*		3.5*	1936–1982	Colle Gnifetti	Wagenbach and Geis (1989)
M		75		100	1985	Capo Cavallo (Corsica)	Bergametti (1989)
M		170-340			?	Eastern Mediterranean	Ganor and Mamame (1982)
E		34			1 event, Nov. 1984	England	Wheeler (1985)
E		7.1			1 event, May 1977	Italian Peninsula	Prodi and Fea (1979)

** A: Alpes; M: Mediterranean; E: Europe.

* Calculated from Ti results and referring to crustal composition of Mason (1966).

obtained in Corsica by Bergametti et al. (1989). All these annual values are in the same range as individual inputs deposited over low altitude areas of western Europe, when particles carried by strong dust storm were washed out (Prodi and Fea, 1979; Wheeler, 1980).

The only one available record concerning high alpine zones was established by Wagenbach and Geis (1989) from 1936 until 1982. Assuming that there was no relevant long-range transport from source areas other than Sahara, a Saharan dust flux of 40 $\mu g\ cm^{-2}\ yr^{-1}$ was inferred from Ti measurements (crustal abundance, $4.4\ ^0/_{00}$) by these authors. However, this Saharan component was calculated after discarding all peaks of obviously local origin and therefore corresponds to the long-range component rather than to the true Saharan component. For that reason, the corresponding Al flux value of 3.5 $\mu g\ cm^{-2}\ yr^{-1}$ should have been not very different from our long-range estimate. As shown in Table 2, this Al value is rather close to our Saharan value for the period 1955–1982, but remains much lower than the long-range input we calculated for the same period. Note that the relative importance of the Saharan flux (eventually underestimated, see Subsection 6.2) with respect to the long-range flux varies along our core, which on the other hand does not cover exactly the same time period as Wagenbach's study. So, the apparent discrepancy between Wagenbach's estimate and ours could either be due to temporal variations of the ratio of Saharan to long-range component, or be linked to snow accumulation rate variability. Indeed, all the Al peaks of remote origin we observed and which corresponds mostly (as Ti) to insoluble species are similar in amplitude (from 200 to 5000 ppb) to values inferred from Ti measurements made by Wagenbach and Geis assuming crustal composition of continental aerosol (from 350 to 2000 ppb). The difference existing between our long-range dust flux and the one published by Wagenbach and Geis is in direct ratio to the difference observed in snow accumulation rate at the two sites (153–158 cm $H_2O\ yr^{-1}$ at Col du Dôme, 20–40 cm $H_2O\ yr^{-1}$ at Colle Gnifetti). Now, particularly at high altitude, strong wind can significantly affect the snow deposition by snow-drifting and wind erosion. At Col du Dôme, the snow accumulation rate varies by a factor up to 3, depending on wind exposure (Pourchet, personal communication). However, as explained before, wind influence should have been weak at our drilling site. At Colle Gnifetti, in contrast, part of the annual layer is blown away especially in winter (Wagenbach and Geis, 1989), but subsequent losses of deposited material remain unknown and therefore were not taken into account in flux calculations. Lastly, because of complex meteorological conditions prevailing over alpine areas, snow accumulation rates and therefore continental fluxes may have been really different at both sites.

The long-range (and Saharan flux) increase, beginning in the early 1970s and strongly marked particularly for Al since 1980, is obvious from Table 2 and Fig. 4, in which local peaks have been discarded for profile smoothing. Wagenbach and Geis (1989), indicated that Ti concentrations were lower from the early 1960s until the mid 1960s than earlier and later.

A similar change in atmospheric concentrations of particles carried to Barbados by the Trade Winds has been described for the 1965–1984 period (Prospero and Nees, 1986) i.e. a marked increase of summer maxima in the early 1970s, becoming sharper in the early 1980s when exceptionally high concentrations were measured. These authors linked their observations to the severe drought afflicting the Soudano-Sahelian zone since 1968, following periods of high (1950–1958) and then normal (1959–1967) rainfall activity. This drought should have contributed to strongly enhanced dust-storm activity in the afflicted area (Middleton, 1985). Those conclusions cannot directly be applied to our study, since the source areas of dust depositing in Alpine regions are probably different (although they are not yet clearly defined, they are probably located in the northern limits of the Sahara rather than in more southern regions), but similar arguments could tentatively be used for explaining the increases we observed. Changes in the frequency of occurrence of suitable meteorological conditions could be an alternative explanation.

Although preliminary, this study has allowed us to point out two main ideas: the first is that Saharan aerosol plays an important and possibly dominant role at high altitude in western Europe with respect to other continental aerosols; the second is that high Ca content of Saharan (and, more generally speaking long-range) aerosol is probably of importance for biogeochemical cycles involving acid neutralisation. Further studies will require a better knowledge of source regions and of meteorological conditions prevailing both over source and deposition areas.

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